Draft Geochemical Model Criteria Support Document for the Dewey-Burdock Project

1. Introduction

1.1. Goals and Need for Geochemical Modeling

Why is geochemical modeling needed?
Geochemical modeling is an important part of the planning, operating, and monitoring activities at the Dewey-Burdock uranium in-situ recovery (ISR) project. It will be used by the site operator and permitting authority to predict changes in the chemistry of groundwater and solids to ensure that the site is behaving as predicted and that underground sources of drinking water (USDWs) are not endangered by ISR activities.

The geochemical model will simulate the movement of fluids (groundwater and lixiviant) and their interactions with solids throughout the project site (i.e., within the orebody and downgradient) to explore the potential for migration of uranium and other metals (e.g., vanadium, arsenic, molybdenum, etc.). Specifically, the geochemical model will simulate various geochemical processes to evaluate the potential for: mobilization of uranium and other metals beyond the aquifer exemption boundaries; the role of natural attenuation in controlling the migration of contaminants; projected concentrations of constituents with maximum contamination levels (MCLs) and NRC-approved alternate concentration limits (ACLs); and potential rebound mobilization of uranium after site restoration. These interactions will be simulated throughout the project's lifecycle. Iterations of and improvements to the geochemical model will incorporate multiple rounds of baseline water quality and solids sampling and the results of monitoring throughout the extraction, restoration, and post-restoration phases (which occur consecutively and concurrently).

The geochemical modeling, coupled with the results of site-specific monitoring (which will provide inputs for future iterations of the model), can help demonstrate compliance with the UIC and NRC permits by showing that the site behaves as predicted through the project's lifecycle; predict possible excursions of injected fluids or contaminated groundwater outside the injection zone (including those that affect USDWs); and support updates to monitoring or the geochemical or conceptual site model (CSM).

The accuracy of the model—and, therefore, its utility as a tool in designing site operations—is improved by the amount and quality of available data on which to base the CSM and geochemical model, including the number of data points and their spatial/temporal distribution. Assessing the gaps in this knowledge supports an understanding of the model's limitations and uncertainties, which can support interpreting the results and designing monitoring programs (baseline and during operational and restoration phases) to fill in data gaps.

How does the geochemical modeling relate to the conceptual site model?
The geochemical model will be based on a CSM that identifies the geology, hydrogeology, and geochemistry of the site, including their baseline conditions as well as during the course of the ISR

project. The CSM helps the model developer identify the relevant site characteristics and geochemical processes to be incorporated into the geochemical model.

The CSM for the site is a representation of the site geologic, geochemical, and hydrogeologic characteristics and the anticipated processes. It describes: the geologic setting of the project site, including the stratigraphy and lithologies of the Inyan Kara Group (i.e., the ore body) and the Graneros Group and Morrison Formation (the upper and lower confining zones); hydrogeologic properties of the site, including static characteristics (e.g., aquifer and confining zone petrophysical properties) and those that will change over the course of the project (such as groundwater flow); and baseline groundwater and sediment geochemistry as well as information on how these change over time. Both the geochemical model and the supporting CSM will be iterative and will be refined as data are collected during ISR/restoration cycles and the post-restoration phase to improve confidence in the results and their usefulness in ensuring USDW protection.

The criteria for the CSM are described in the *Draft Criteria for Development of a Conceptual Site Model of the Dewey-Burdock Project* (CSM criteria document) and are accompanied by the *Draft Conceptual Site Model Criteria Support Document for the Dewey-Burdock Project* (CSM support document).

1.2. What is Geochemical Modeling?

A geochemical model represents the geochemical information in the CSM mathematically by means of a modeling code (i.e., computer program) that includes the equations governing the geochemical and biogeochemical processes that are anticipated to occur at the site. The processes typically simulated in a geochemical model include aqueous speciation (the form of a constituent in solution), dissolution and precipitation of minerals, ion exchange, adsorption and desorption of constituents from mineral surfaces, redox reactions, and biogeochemical processes. (Zhu and Anderson, 2002; Nordstrom, 2003; Appelo and Postma, 2005).

There are several types of geochemical models: aqueous speciation-saturation, forward (reaction path), inverse (mass balance), and reactive transport. Speciation-solubility modeling calculates the concentrations and speciation of species in solution and whether the solution will dissolve or precipitate minerals (saturation indices). Forward modeling uses selected reactions and other assumptions to predict water composition. Inverse modeling uses data on water and solids chemistry to determine the reactions that have taken place (Nordstrom, 2003). Inverse modeling can also be used with experimental data to estimate parameters (e.g., sorption parameters) or to elucidate which processes will be important in controlling water chemistry at the site.

Because the groundwater composition is affected by interactions with the aquifer solids during flow, geochemical modeling codes are often coupled to the simulation of fluid flow to model what is termed reactive transport. The transport component of reactive transport may entail fluid flow in one, two, or three dimensions (1-D, 2-D, or 3-D). Although the term "geochemical model" will be used in this document, it is assumed that any modeling approach for the Dewey-Burdock site will employ numerical methods and entail reactive transport. EPA: Do you want a paragraph on some of the common programs used for geochemical and reactive transport modeling?]

A geochemical model that incorporates transport allows exploration of assumptions and the estimation of future conditions such as changes in groundwater chemistry over time, how quickly a contaminant

will move through the subsurface, when it will reach a boundary (e.g., aquifer exemption boundary), what factors control contaminant migration, and what changes may take place in the geochemistry of the solids.

It is important to remember that the computer program used for modeling is not the model; the model is the representation of the system as developed by the modeler, including all of the selected assumptions for depicting the geologic/hydrogeologic setting and selecting the geochemical species, data, reactions, and databases. There are always uncertainties and inaccuracies in modeling stemming from data quality and quantity as well as uncertainties inherent in the modeling process itself, including databases used by the program. Nonetheless, modeling is a valuable tool for assessing risk, estimating contaminant fate and transport, and supporting and defending permitting decisions (Newman, 2018).

The main components needed to conduct modeling are the selection of a modeling program, the thermodynamic and kinetic data and equations needed to simulate the geochemical processes, and site-specific data (i.e., water quality data, solids characterization, and experimental data if needed to estimate parameters for processes). All modeling has limitations. The main limitations of geochemical modeling are the amount of available field and laboratory data, whether the data are representative of all areas of the project site, the quality of the data, and any limitations or uncertainties in the thermodynamic and kinetic data used in the modeling code. Increasing the amount and quality of the site-specific data that drive the model assumptions and provide inputs will decrease uncertainty and improve the accuracy of and confidence in the model results.

Additional steps once the model has been framed are calibration to parameterize the model and sensitivity analysis to evaluate uncertainty. Model calibration is the process of iteratively adjusting model parameter estimates to improve the fit between model output and an independent set of measured data (i.e., the results of groundwater monitoring at the site). Based on the level of agreement between model outputs and the independent data, the model is either considered successfully calibrated for application or model input and parameters are revisited for further calibration. Although the level of calibration undertaken will depend on modeling objectives and data availability, all modeling efforts must contain some assessment of the quality of model outputs in order to guide model application.

A sensitivity analysis is a systematic and methodical investigation of the effects of alternative model parameter values on simulation results. The application of a sensitivity analysis as part of geochemical modeling is typically completed to enhance the conceptual understanding of the modeled system, identify parameters of greater interest for model calibration, and to better understand how uncertainties in parameter values may translate to uncertainty in model outputs. A sensitivity analysis of model parameters is a critical part of the development of a geochemical model.

1.3. Resources on Modeling

Geochemical modeling, including reactive transport, is a complex discipline; it is only discussed at a general level in this document in order to highlight important aspects of the process, including data needs and sources of uncertainty, as they relate to modeling of the Dewey-Burdock site. For detailed overviews of geochemical modeling, a number of resources are available. A few examples include: Nordstrom (2003); Zhu and Anderson (2002); Appelo and Postma (2005); Alpers and Nordstrom (1999);

and Nordstrom and Campbell (2014). For background information on groundwater chemistry, see Stumm and Morgan (1981) or Drever (1988). Guidance on evaluating geochemical modeling at mining sites was developed by the Nevada Division of Environmental Protection (Newman, 2018). For the transport aspects of reactive transport modeling, EPA (1994) has produced a technical guide for groundwater model selection at sites contaminated with radionuclides; it provides discussion of modeling codes, including numerical models. Reilly and Harbaugh (2004) developed guidelines for evaluating groundwater flow models, and a USGS report by Franke et al. (1987) provides an overview of boundary and initial conditions related to modeling of groundwater flow.

1.4. Previous Modeling at In-Situ Recovery Sites

Geochemical modeling has been used successfully at several other in-situ recovery sites; this provides a reasonable precedent for modeling at the Dewey-Burdock site. One such site is the Smith Ranch-Highlands uranium project, an ISR mine in Wyoming. Geochemical modeling has been used to predict post-operational groundwater restoration at several stages of the project. The Smith-Ranch Highland A-Wellfield was mined for uranium from 1988 to 1991 using a lixiviant consisting of groundwater, carbon dioxide, and oxygen. Restoration was completed in 1998, but a 15-year natural attenuation monitoring plan included groundwater flow modeling with MODFLOW and PHREEQC geochemical modeling. Data collected semi-annually from four monitoring wells placed strategically around the wellfield supported this modeling. Over a year of stability monitoring was needed after restoration was completed in order to sufficiently characterize long-term attenuation trends (U.S. EPA, 2011).

More recently, a one-dimensional PHREEQC groundwater transport model was developed for Mine Unit 1 at the Smith Ranch-Highland project after groundwater restoration was completed in 2014. Six different geochemical models were developed for six preferential flow paths, which were identified as part of the conceptual site model. Each used PHREEQC (version 3), the Enchemica.R1.dat geochemical database, and 1 m cell sizes. The model also incorporated monitoring well data collected in the years following restoration. Overall, it predicts that natural attenuation will return downgradient groundwater to a state similar to pre-ISR operations (Enchemica, 2018).

In an example of iterative comparison of geochemical modeling results with monitoring well data to assess current restoration efforts and recalibrated to account for adjustments, operators at the Beverley Four Mile uranium project in southern Australia designed a model that combines groundwater transport in a dual porosity approach with PHREEQC geochemical calculations. After initially calibrating the model with laboratory batch and column tests, they found that natural attenuation, enhanced with a groundwater sweep, has the potential to sufficiently restore the site. The model will continually be compared with monitoring data, however, in order to determine whether additional restoration techniques will be necessary. The Beverley Four Mile project geochemical model also used higher resolution cell sizes, depicting the uranium ore bodies in small patches rather than in a large block (Jeuken et al., 2009).

Previous geochemical modeling has already been completed at the Dewey-Burdock site. A team at the U.S. Geological Survey (USGS) designed a one-dimensional column with cells 5 meters in length and used PHREEQC for the geochemical calculations. They found that their predictions were highly sensitive to subtle changes in groundwater geochemistry, particularly changes in pH and in calcite equilibrium. The authors noted that any model of restoration at the site should take into account the differing

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geochemistries of the groundwater at baseline conditions, during restoration, and as groundwater moves through the site during the restoration phase (Johnson and Tutu, 2016). For this model, however, only three downgradient core samples were available for analysis. Any future geochemical modeling would need additional samples to better characterize the site, especially given its heterogeneity. Modeling will also need to draw on the latest thermodynamic data for uranium speciation (Johnson and Tutu, 2016). Overall, new geochemical models can use the work done by USGS as a starting point for creating more refined simulations of groundwater dynamics at Dewey-Burdock.

1.5. Purpose of Support Document

The purpose of this document is to provide background information about and considerations for the development of a geochemical model that incorporates site-specific geological, hydrogeological, and geochemical characteristics of the Dewey-Burdock site. The geochemical model will simulate the geochemical processes that are expected to occur during the life cycle of the ISR project. The remaining sections of this document present background information and considerations for:

- Setting up the model to represent site-specific geologic and hydrogeologic features and other inputs such as boundary and initial conditions and expected geochemical reactions (Section 2);
- Thermodynamic and kinetic data and handling of sorption (Section 3);
- Groundwater and solid phase geochemical data to accurately predict the groundwater-solids interactions that govern the speciation and mobility of uranium and other metals that may be mobilized during the ISR project (Section 4);
- Calibration of the geochemical model through a comparison of model outputs with field samples and laboratory experiments (Section 5); and
- Sensitivity analyses to understand which inputs and settings contribute most to output variability and, therefore, require greater attention during model calibration (Section 6).

2. Background and Considerations for Model Setup

A key part of developing an accurate model is the assumptions made for model setup. These include the establishment of the model domain and the representation of the geologic, hydrogeologic, and geochemical characteristics; the duration of the simulation and time steps; selection of constituents and reactions to represent; and the boundary and initial conditions. This section provides a brief overview of basic considerations; the reader is referred to the resources listed in Section 1.2.1.

Domain boundaries (i.e., the space being modeled) may be defined by natural features such as geologic structures, water bodies, or natural divides in groundwater flow. In general, the boundaries of the domain should be sufficiently far from areas of concern to minimize errors related to the setting of boundary conditions. As described in the geochemical modeling criteria document, the model domain should be defined to represent all areas of the Dewey-Burdock site relevant to the simulation of groundwater flow and geochemical processes associated with ISR. This includes representing the upgradient area, the extraction zone, and downgradient areas, including a margin beyond the aquifer exemption boundary to predict any unanticipated excursions outside of the injection zone or exempted aquifers.

The assignment of geologic and hydrogeologic characteristics to parts of the model domain will be based on information gathered to develop the CSM. Additional discussion regarding representation of the geologic and hydrogeologic properties is presented in Section 2.1 and Section 2.2. Other inputs, such as boundary and initial conditions and selection of constituents and reactions, are discussed in Section 2.3.

2.

2.1. Background and Considerations for Geologic Criteria

Sufficient, detailed understanding of site geology and stratigraphy, including the depths and lithologies of the injection formation and the upper and lower confining zones is important for simulating the movement of native groundwater and injected fluids within the formations. The computational simulations conducted for modeling are only an approximation of the actual system, so it can only predict system behavior if the complexities of the subsurface (which are, ideally based on site-specific data collected at the site or, alternatively based on relevant literature) are captured in the computational inputs.

A complete geologic dataset will include detailed information about the lithology of the injection zone, the confining zones, and any other permeable formations, including USDWs, that may be affected by vertical fluid migration. For the Dewey-Burdock site, this would include the Inyan Kara Group injection zone, the Graneros Group and Morrison Formation (the upper and lower confining layers), and the surrounding stratigraphic units such as the Quaternary alluvium or the Unkpapa Sandstone. For more information about the geologic data needed for these units, please see the CSM Criteria Document and the CSM Support Document. Having site-specific data from the Dewey-Burdock project area is critical to defining the system boundary and initial conditions. Without these data, the geochemical model will be limited to rudimentary calculations of groundwater flow and contamination (U.S. EPA, 1994).

Much of the information needed to develop the model is available, either in the UIC permit application itself (Powertech, 2011), or previous work by USGS (see, e.g., Johnson and Tutu, 2016). More samples, however, will likely need to be collected from the site to reduce the model's uncertainty. A reliable dataset that is representative of all parts of the field, both upgradient and downgradient from the ore zone can mitigate this uncertainty.

In a reactive transport model, the geologic information will be represented by a system of one-, two-, or three-dimensional cells. The model represents the geologic system in a series of discretized cells that each represent specific geologic characteristics. If a cell is defined as 1 m^3, for example, then that cell will represent an area of rock in the subsurface that is 1 m^3 and will have the geologic properties of that rock as they are defined in the dataset. The precise manner in which the site geology is represented in the model is up to the modelers, so long as it is designed in a way that is consistent with site data and will generate useful results. See [REF _Ref10814187 \h] for an example of a 3-D model representation showing discretization of the geologic formations at a hypothetical site.
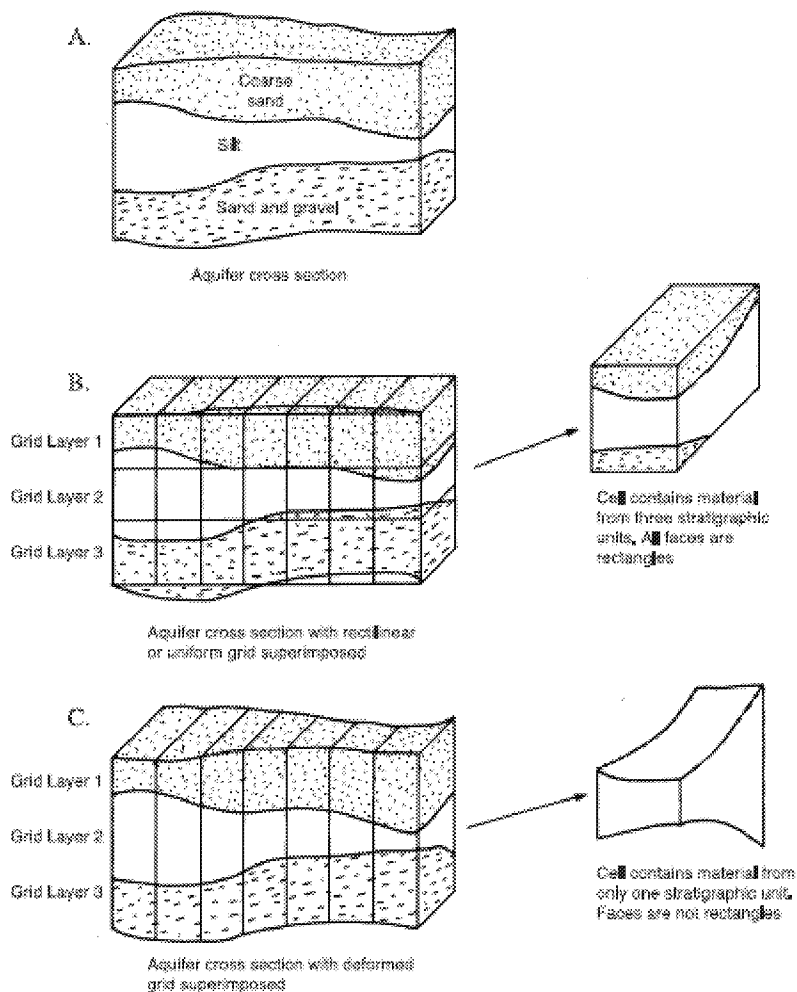
Figure [SEQ Figure * ARABIC]. Schemes of vertical discretization for (A) aquifer cross section, (B) aquifer cross section with rectilinear or uniform grid superimposed, and (C) aquifer cross section with deformed grid superimposed. Source: Reilly and Harbaugh (2004) Note: Permission not obtained.

2.2. Background and Considerations for Hydrogeologic Criteria

Site-specific hydrogeologic information about the Inyan Kara Group and the confining zones based on data collected from within the proposed extraction area, upgradient of the proposed extraction area, and downgradient, will support assumptions for an accurate geochemical model. As with the geologic data, the accuracy of the model's predictions will be improved with a greater number (i.e., in detail/density) and quality of the site-specific data inputs.

A complete dataset will include detailed information about the hydrology of the injection zone, the confining zones, and any other permeable formations that may be affected by vertical fluid migration. For the Dewey-Burdock site, this would include the Inyan Kara Group injection zone, the Graneros Group and Morrison Formation confining layers, and the surrounding stratigraphic units such as the

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Quaternary alluvium or the Unkpapa Sandstone. Within the Inyan Kara Group are two different injection zones, the Chilson Member of the Lakota Formation and the Fall River Formation, separated by the impermeable Fuson Shale. The geochemical model will need to represent this complete stratigraphic column. For more information about the hydrogeologic data needed for these units, please see the CSM Criteria Document and the CSM Support Document.

Hydrogeologic data collected from the Dewey-Burdock site can include porosity and lithology from well logging; hydraulic conductivity, storativity, and transmissivity from aquifer testing; potentiometric data; downhole pressure data; and any other relevant test- or field-based observations (Franke et al., 1987). Much of this information is available in either the UIC permit application (Powertech, 2011), or previous USGS work (see, e.g., Johnson and Tutu, 2016). It is important to note, however, that groundwater flow direction will be in flux during the ISR project, as water is injected and extracted from the field, and then during restoration and the reestablishment of natural flow patterns in consecutive and concurrent processes. Thus, the model will need to predict these changes in groundwater flow but will also be recalibrated both during and after ISR operations, with updated inputs based on data gathered during monitoring.

Because of the complexity of the subsurface, the geochemical model will need to be designed to mitigate uncertainties. First, decisions will need to be made regarding how to handle variability in porosity and permeability measurements. There may or may not be agreement between log-based and core-based measurements, and porosity logs may have been used to generate permeability estimates. Second, there may be localized heterogeneity throughout the field. The modelers must decide whether to use average values or ranges of values and whether to develop statistically-based porosity and permeability distributions within the injection formation. Lastly, the resolution of the model, i.e., the size and dimensions of the cells, must be selected based on the needs of the project and planned application of the modeling results and documented. Ultimately, issues of uncertainty are dependent on having a sufficient density of data throughout the project area.

2.3. Other Inputs

Boundary conditions are described by U.S. EPA (1994) as "...the conditions the modeler specified as known values in order to solve for the unknowns in the problem domain." For geochemical characteristics, this can include the mineralogic composition of the solids and the groundwater chemistry at the boundaries of the model domain. Hydrogeologic boundary conditions describe the groundwater flow in and out of the system (see Franke et al., 1987 for discussion of types of hydrogeologic boundary conditions).

Initial conditions are those conditions in the model domain at the beginning of the simulation. These include information such as the initial distribution of constituents of concern, the mineralogy of the solids, initial water chemistry, pressure and temperature, and redox conditions. Hydrogeologic data, such as hydraulic head or flow volumes also need to be specified throughout the domain (Reilly and Harbaugh, 2004). Boundary conditions and initial conditions are often discussed together. Both require adequate data, collected as part of a site characterization and multi-stage baseline monitoring program, to represent the site as accurately as possible.

Other choices that the modeler must make include specifying the chemical species to include in the model, the minerals present, and the anticipated geochemical reactions. It is not possible to specify every reaction that can take place in the system, but the model should at least include processes that have a significant effect on the overall water chemistry as well as trace elements of concern. For forward modeling (reaction path), decisions about which processes to represent in the model should be based on the most important site characteristics and the needs of the project. For the Dewey-Burdock site, geochemical processes to specify in the model should include those identified in the CSM as being significant for the mobilization and retardation of uranium and other constituents of concern, including those with MCLs or ACLs. Where possible, the oxidation state of a species should be specified. Other decisions to be made by the modeler include whether to include the kinetics (rates) of reactions or if it is warranted to assume local equilibrium (see Section 3).

3. Background and Considerations for Criteria Related to Thermodynamic and Kinetic Data

Along with selecting the components and reactions to represent in the model, the modeler will need to determine whether to allow the program to assume local equilibrium or include reaction kinetics. The thermodynamic equations that serve as the foundation for geochemical modeling represent processes that reach equilibrium (stable state, with lowest energy). Although the larger system is dynamic and not in equilibrium, equilibrium may be assumed at a small-scale, local level. In some systems, however, the geochemical processes may be controlled by the rates at which reactions take place (Appelo and Postma, 2005). The modeler will need to consider, based on available information about the site, whether kinetics should or can be represented (Newman, 2018) . Sections 3.1 and 3.2 briefly introduce some of the relevant considerations and types of information. These topics are complex, with decades of literature covering the theoretical bases, development of data, and implementation in geochemical models and modeling programs (e.g., Nordstrom, 2003; Zhu and Anderson, 2002; Appelo and Postma, 2005; and other sources noted in Section 1.2.1). Also, although multiple geochemical processes will be active at the Dewey-Burdock site (mineral precipitation/dissolution; aqueous complexation; redox changes; sorption), Section 3.3 deals explicitly with sorption because of its importance in controlling the mobility of U(VI).

3.

3.1. Thermodynamic Data

Thermodynamic data are central to the computations performed in geochemical modeling. Modeling programs have built in thermodynamic databases containing the data (thermodynamic properties and equilibrium constants) needed to simulate equilibrium processes. The thermodynamic data for the aqueous and solid species involved have a significant effect on model output; differences in the equilibrium constants used may mean the difference between predicting that a mineral is thermodynamically favored to dissolve vs. precipitate. The accuracy, completeness, and internal consistency of these databases is key for accurate modeling results. There are several compilations that are commonly used, including those used by the USGS and USEPA codes (Nordstrom et al. (1990) and

Nordstrom and Munoz (1994)). See Nordstrom (2017) for a review of thermodynamic databases and modeling programs.

The limitations and challenges that can affect the model results can include a lack of data or poor data for some species or minerals. Newman (2018) calls attention to the potential for internal inconsistencies in some databases. If species important for a system are not represented in the database or there are newer data that the modeler determines are appropriate to use, the data can be edited. For uranium, Guillaumont et al. (2003) presents the fifth report in a series of critical reviews of thermodynamic data published by the OECD Nuclear Energy Agency; the thermodynamic data in this report should be considered for inclusion in the thermodynamic database that is used for the modeling of the Dewey-Burdock site. Also, Muhr-Ebert et al. (2019), developed a database building on the Guillaumont et al. (2003) report, including modifications for calcium and magnesium uranyl carbonates.

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2.1.

2.1.1.   Activity Coefficient Models

Thermodynamic equilibrium constants are defined as if the fluid were infinitely dilute. To model equilibrium reactions in natural waters, however, an activity coefficient must be applied to the concentration of each constituent to compensate for interactions among ions in the water (e.g., Appelo and Postma, 2005). There are a few equations for calculating activity coefficients. The Debye-Huckel, Davies, and B Dot equations are most suitable for relatively dilute waters (Stumm and Morgan, 1981; Helgeson, 1969). For high ionic strength waters, the specific ion interaction theory (SIT) (i.e., Pitzer equations) is an appropriate approach (e.g., Pitzer, 1979; Harvie and Weare, 1980; Harvie et al., 1984; Plummer et al., 1988), although it is applicable to fewer constituents.

Selecting the best activity coefficient model for the Dewey-Burdock site may be complicated by changing fluid composition (ionic strength and specific constituents) due to injection of the lixiviant, mixing of the lixiviant with native groundwater, and introduction of restoration fluids. It is also important to verify that there are data for the constituents of concern. [EPA: we need to verify which equations are suitable for U, V, and other metals as well as check the ionic strength of the groundwater.]

3.2. Kinetic Data

If local equilibrium cannot be assumed in forward modeling, use of kinetic expressions for reactions should be considered. The limitations in successfully modeling kinetics can include a lack of data to determine the most appropriate or acceptable formulas for reaction rates and parameterize them by estimating the coefficients for the formulas. Geochemical modeling programs can include built-in rate laws, the formulas that describe the relationship between the reaction rate and constituent concentration (also factoring in temperature). If the geochemical program does not include the appropriate rate law for a reaction, user-defined rate laws can be entered.

Built-in capabilities in modeling programs can make use of existing datasets, such as the compilation of rate parameters for water-mineral interactions by Palandri and Kharaka (2004), which references an extensive number of studies. If reactions relevant to the Dewey-Burdock site, such as those involving uranium and other metals, are not adequately covered in existing site-specific data, information derived from recent literature that is based on analogous sites can be incorporated into the model. Laboratory column experiments at conditions representative of the site can provide information on reaction rates and improve the site-specificity of the model (e.g., Ben Simon et al., 2014).

3.3. Sorption

Transport of U(VI) in groundwater is known to be controlled by adsorption to mineral surfaces (e.g., Johnson et al., 2016a). Therefore, optimal simulation of sorption at the Dewey-Burdock site is a key aspect of estimating the potential for transport of U(VI) where oxidizing conditions exist. Johnson and Tutu (2013) emphasized the need for good site-specific information, including iron in the aquifer solids, and experimental work to effectively model uranium adsorption and predict downgradient concentrations at ISR sites.

Empirical representations of sorption (e.g., Kd, Langmuir isotherms and possibly Freundlich isotherms; Drever, 1988) can typically be used in geochemical modeling programs. However, because the groundwater chemistry at the Dewey-Burdock site is anticipated to change spatially and temporally, a surface complexation model (SCM) approach may be more appropriate. Laboratory experiments, which are described in the CSM criteria document, should be structured to provide the necessary data to support the chosen approach for modeling sorption.

The SCM approach describes the formation of chemical species at the mineral surfaces (surface complexes) as constituents in the water interact with the mineral surfaces (Davis et al., 2004). This is advantageous because the role of aqueous complexes is incorporated in the complexation reactions At the Dewey-Burdock site, the formation of uranyl-carbonate and calcium-uranyl-carbonate complexes will contribute to uranium solubility and reduce sorption (e.g., Dong and Brooks, 2006). The formation of surface complexes is described as an equilibrium process. It is appropriate for systems with changing groundwater geochemistry and can be incorporated into reactive transport models.

A rigorous SCM approach addresses the formation of complexes at the surfaces of specific mineral types. Therefore, a significant amount of thermodynamic and mineral characterization data is needed. Alternatively, a somewhat simpler SCM approach, generalized composite (GC) treats the sediment as a whole as far as characterization (Davis et al., 2004). This makes site-specific data easier to obtain because experimental work does not require specific minerals to be separated out. Johnson et al. (2016a) conducted laboratory batch experiments specific to the Dewey-Burdock site and estimated sorption parameters using this approach. The authors note, however, that they were working with a limited number of samples and also recommend that future work look at the role of kinetics in sorption at this site.

4. Background and Considerations Related to Use of Site Geochemical Data

The accuracy and completeness of the water quality and solids analyses affect the accuracy of the model. If important constituents were either not analyzed or have problematic analyses, the accuracy or certainty of the modeling result may be affected. For example, either the resulting predictions of contaminant mobility and water quality changes will have increased error, or certain processes may not be able to be properly represented. Water quality analyses and the associated quality assurance (QA) needs at the Dewey-Burdock site will be guided by provisions in the NRC license and draft Class III UIC permit (U.S. EPA, 2017). Solids analyses should be guided by project needs and comprehensive enough to provide information about initial conditions and processes to be modeled. Sections 4.1 and 4.2 discuss considerations regarding the quality and utility of groundwater and solids analysis.

4.

4.1. Groundwater Quality Data

Good quality groundwater analyses are crucial for obtaining valid modeling results. A common way to initially screen groundwater analyses is to evaluate the charge balance, the balance of positive and negative charges between the measured concentrations of cations and anions. Theoretically, the charge balance should be zero. However, because water analyses inherently have some uncertainty associated with the analytical methods, there will likely be some charge imbalance (Zhu and Anderson, 2002). If the charge balance is beyond an acceptable range ($\pm$ 10% is a common criterion), then a significant constituent may not have been analyzed, or the sample handling or analysis was faulty.

Note that if the charge balance is calculated using only the ionic species of each element, the charge imbalance may be spuriously high. The geochemical modeling program should perform a speciated analysis to represent the forms in which the constituent would be present in the water.

If a charge balance problem cannot be remedied by, for example, a repeat analysis or use of other data, geochemical modeling programs can force a charge balance by changing the composition of one of the constituents. Chloride is sometimes used for this, especially if it already a significant constituent (Zhu and Anderson, 2002).

Even if the charge balance is acceptable, there may be problems with the analysis if two errors balance each other out. For this reason, data should be evaluated against historical data if possible and ion ratios evaluated to identify possible outliers (Stratus Consulting et al., Undated) . As always, the data should make sense intuitively given what is known about the geology, history of activities, and other information about the site. Field measurements for temperature, pH, dissolved oxygen, Eh, and alkalinity should be available to accompany the analytical results from sampling.

4.2. Solid Phase Geochemical Data

It is important to represent the aquifer solids at the Dewey-Burdock site as completely and accurately as possible when developing the geochemical model to improve the accuracy/reduce the uncertainty of

predicted water-solids interactions. The minerals identified during development of the CSM should be represented in the geochemical model, with attention paid to variability in mineralogy and oxidation state in the different portions of the project site. Information on aquifer solids may have been obtained through several methods (e.g., X-ray diffraction, polarized light microscopy, scanning electron microscopy, electron microprobe, sample digestion and analysis, Brunauer–Emmett–Teller (BET) surface area). These data should support the modeling needs. For example, X-ray diffraction identifies mineralogy, but will not identify trace minerals that may control the concentrations of trace elements if dissolution occurs. Mineral composition (e.g., by electron microprobe) may be needed to identify trace contaminants in solid solution that may be mobilized if dissolution occurs (e.g., arsenic in pyrite). Whole sample digestion and analysis (e.g., via Inductively coupled plasma mass spectrometry, ICP-MS) may help in identifying variability in the concentrations of constituents of concern. Data from X-ray absorption near edge structure (XANES) analysis will provide information on uranium oxidation state. Scanning electron microscopy may help characterize grain coatings of iron oxides that play a major role in adsorption. BET analyses may be needed as part of solids characterization for surface complexation modeling. It is not a given that all of these analyses are necessarily needed for all samples, but the proper analyses of samples collected throughout the Dewey-Burdock site should be planned to provide the necessary data for developing both the CSM and the geochemical model. See the CSM Criteria Document for additional discussion regarding solids characterization.

5. Background and Considerations Related to Geochemical Model Calibration

Model calibration is the process of iteratively adjusting model parameter estimates to improve the fit between the model output and an independent set of measured data (referred to hereafter as observations) that are based on sampling and analysis of groundwater collected at the site or experimental results. Based on the level of agreement between model outputs and the observations, the model is either considered successfully calibrated or model input and parameters are revisited for further calibration. Calibration typically involves a combination of manual adjustment of parameters based on perceived graphical agreement between modeled and observed data and goodness-of-fit statistics (often referred to as a trial-and-error approach) and the use of automated methods termed inverse models that attempt to optimize parameter values using statistical techniques and many iterative model runs. A comprehensive review of groundwater model calibration is provided in Hill and Tiedeman (2006).

The calibration process begins by identifying the model output variables that will be compared to observations in order to guide parameter adjustment. Factors to consider during this step include an evaluation of which outputs are most relevant to the intended application of the model and the number and quality of observations available for each output term. For the Dewey-Burdock geochemical model, outputs are expected to support project planning and to help evaluate whether there may be mobilization of uranium and other constituents beyond the aquifer exemption boundaries or rebound mobilization of uranium after site restoration, and the role of natural attenuation in controlling the migration of contaminants. Results may help guide adjustments in operational or monitoring practices to protect USDWs. Key outputs for application therefore include groundwater concentrations of

uranium and other constituents of concern for drinking water quality (e.g., MCLs, ACLs) at various locations throughout the system and over time.

After outputs are selected for calibration, observations of the output terms must be collected and compiled. For geochemical models, observations could include field samples of groundwater quality or measurements from laboratory experiments of sorption reactions conducted with representative subsurface solid material and liquids. Field samples of groundwater quality in and around the Dewey-Burdock site have been collected as part of site permitting and will continue to be collected under direction of the site monitoring plan. The collection of water quality data during ISR injection, extraction, restoration, and post-restoration will provide observations against which model performance can be evaluated during each phase and model settings adjusted accordingly.

Hill (1998) notes that since groundwater model parameters can be highly spatially or temporally variable, the number of parameters to calibrate to could theoretically be infinite. In contrast, a limited number of observations are available for evaluating and adjusting a model. The discrepancy between the number of observations and the number of parameter values requiring adjustment is addressed in two ways. First, assumptions on the homogeneity of the system are applied so that parameter values only reflect major differences in hydrogeochemical conditions. Second, a sensitivity analysis is commonly completed as part of model calibration to identify which parameters are most relevant to the outputs of interest. Previous efforts to develop geochemical models for uranium ISR sites, including the Dewey-Burdock site, have demonstrated high sensitivity of uranium transport predictions to surface sorption parameters (site densities and equilibrium constants), groundwater pH, calcite equilibrium, and alkalinity (Curtis et al., 2006; Johnson and Tutu, 2016b; Ben Simon et al., 2014). Sensitivity analysis concepts and considerations are described in further detail in Section 6 of this report.

Quantitative calibration requires the definition of numerical measures of model performance. Newman (2018) notes that although some geochemical modeling efforts analyze the mathematical agreement between outputs and observations, "there is currently no widely-used method to evaluate geochemical predictions mathematically and quantitatively." Example geochemical modeling studies have considered various performance measures, including the sum-of-squares weighted residual (Johnson et al. 2016b), root mean squared weighted error, mean percentage error, and coefficient of determination (Karlsen et al., 2012).

In addition to the selection of one or more performance statistics, modelers must determine whether a manual trial-and-error or automated inverse modeling approach will be used to identify the parameter values that optimize model performance. Hill (1998) notes that the benefits of automated inverse modeling include a clear determination of parameter values that produce the best possible fit to the available observations; diagnostic statistics that quantify the quality of calibration and data shortcomings and needs; inferential statistics that quantify the reliability of parameter estimates and predictions; and the identification of issues that are easily overlooked during non-automated calibration. Drawbacks include insensitivity of model performance measures to parameter changes due to insufficient observation data; non-uniqueness of optimal parameter sets; and instability of model outputs to slight changes in parameter values. These issues must be considered by modelers during the data collection process and throughout model calibration.

6. Background and Considerations Related to Sensitivity Analyses and Uncertainty Prediction

A sensitivity analysis is a systematic and methodical investigation of the effects of alternative model parameter values on simulation results. The application of a sensitivity analysis as part of geochemical modeling is typically completed to enhance the conceptual understanding of the modeled system, identify parameters of greater interest for model calibration, and better understand how uncertainties in parameter values may translate to uncertainty in model outputs. This process can provide insight on the relevance for application at the site and permitting decisions). There is no single universal approach to sensitivity analysis. The methodology and level of complexity will vary from one study to the next depending on the model configuration and modeling objectives (Newman, 2018; Pianosi et al., 2016).

Pianosi et al. (2016) describe several different attributes of a sensitivity analysis method. These include:

- Local versus Global. A local sensitivity analysis evaluates the variability of model outputs within a specified range around the initial value of a model parameter. In contrast, the entire theoretical range of the parameter is analyzed under a global sensitivity analysis.
- Quantitative versus Qualitative. Quantitative methods use numeric measurements to characterize the relative influence of a model parameter on model output. A qualitative approach relies on visual inspection of model outputs with varied parameter values
- One-at-a-Time versus All-at-a-Time. In the one-at-a-time method, the sensitivity of a single parameter is evaluated while other parameters are kept at fixed values. The all-at-a-time method allows all parameters included in the sensitivity analysis to vary simultaneously, thereby providing information on the interactive effects of the parameter set.

The above attributes can be combined within a single modeling study. For example, a sensitivity analysis may include a local analysis of one parameter and a global analysis of another and may start with a qualitative visual inspection of results before initiating a quantitative method.

Examples of model processes to consider for a sensitivity analysis of a mine site geochemical model are listed in Newman (2018) and include mineral solubility, sorption reactions, gas exchange, fracture depth and density, groundwater flow rates and influent chemistry, and effective porosity. Other studies provide further insight into model parameters to include in the sensitivity analysis of the Dewey-Burdock geochemical model. Curtis et al. (2006) present a sensitivity analysis of a surface complexation model to describe uranium adsorption that evaluated flow parameters (hydraulic conductivity, recharge, porosity, and dispersivity), sorption parameters (equilibrium constants and site densities), and the chemistry of both contaminated recharge water and background groundwater (uranium, alkalinity, and pH). Sensitive parameters included recharge and porosity, site density, and alkalinity and uranium concentration.

Ben Simon et al. (2014) included a sensitivity analysis of a laboratory-scale model of the ISR process to evaluate how uranium recovery is influenced by lixiviant acidity, subsurface ore zone mineralogy, and the flow rate between injection and recovery wells. Changes in the lixiviant acidity, flow rate, carbonate content, and iron oxide content were found to increase subsurface acid consumption following injection and/or increase the volume of recovered leachate required to satisfy a target level of uranium recovery.

Johnson and Tutu (2016) present results of a sensitivity analysis of a reactive transport model of post-restoration uranium transport from the ISR zone at the Dewey-Burdock site. After establishing a calibrated base model, alternative simulations were completed with calcite equilibrium, varied pH levels throughout the system, and different pH levels in the restored zone versus background groundwater. Results showed greater uranium mobility with calcite equilibrium and increases in pH. Together, these studies provide a starting point for understanding important parameters in the Dewey-Burdock geochemical model that should be given greater attention during the data collection, model set up, and model calibration stages.

A sensitivity analysis can provide an understanding of how uncertainties in parameter inputs translate to uncertainty in model outputs. Geochemical models are typically deterministic, producing fixed and precise outputs for a given set of parameter values. Although not quantified within the model formulations, model outputs contain an inherent level of uncertainty. The uncertainty in model output stems from uncertainty in the parameter values specified by the user and in model structural uncertainty, which can be described as inadequacies in the representation of real-world processes by the model. The uncertainty in model parameters and outputs can be characterized with confidence and prediction intervals. Hill (1998) notes the distinction between these as, "confidence intervals are intervals in which the true parameter value or true predictive quantity is likely to occur with some specified probability. Prediction intervals differ from confidence intervals in that they include the effect of measurement error. Prediction intervals need to be used if the intervals are to be compared to measured values and are most commonly constructed for simulated predictions."

Evaluating and reporting on model uncertainty is important for informing how model findings fit into decision making processes (i.e., permit conditions). Ye (2013) notes the challenges of quantifying uncertainty in reactive transport models relative to groundwater flow and nonreactive transport modeling, highlighting the nonlinear model structure and complex responses of model outputs to varied input values. Johnson and Friedel (2009) point to three potential methods for quantifying uncertainty in geochemical models of ISR sites, including the simulation of multiple conceptual models, the use of a Monte Carlo approach to iteratively produce many output values from randomly selected sets of parameter values, and an inverse modeling approach that calculates prediction uncertainty as part of sensitivity analysis and calibration optimization. Example applications of these approaches can be found in Crescenti et al. (1996), Runkel et al. (2011), Karlsen et al. (2012), and Ye (2013).

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